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(54) Title: SOLID COSMETICS HAVING MOISTURIZING EFFECT (57) Abstract The invention relates to a pressed cosmetic composition which has a high level of moisturizing agent and adequate cake hardness, containing 70-99 % powder colorant and 1-30 % of a binder base containing moisturizing agent and nonionic surfactant which is in liquid or paste form at 25 °C. The binder base also meets at least one of the following requirements: the nonionic surfactant is at least one fifth by weight of the moisturizing agent; or the binder base is a lipophilic gel having a resistivity of at least 10,000 omega.cm at room temperature.		

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Solid cosmetics having moisturizing effect

TECHNICAL FIELD

The present invention relates to a pressed powder cosmetic composition which comprises a high level of moisturizing agent and has adequate cake hardness. The composition is particularly useful for make-up cosmetics such as powder foundation, eyeshadow, blusher, face powder, and powder lip.

BACKGROUND

Cosmetic compositions containing a high level (70% or more) of powder colorants are known as pressed powders and are typically made by spraying a binder base consisting essentially of oily materials onto the powder colorants and pressing into a cake. These pressed powder cosmetic compositions are generally known to cause drying of skin due to moisture take-up or absorption by the powder colorants from the surface of the skin. However, when an effective amount of moisturizing agent is incorporated in a composition with high level of powder colorant, the moisturizing agent agglomerates the powder colorants. Consequently, the product which has then been pressed into a cake becomes unacceptably hard. It is known in the cosmetic art that pressed cosmetic cakes which are too hard have a solid and slippery surface, and thus are difficult to take-up with an applicator, and also show a negative shiny, glossy appearance (known in Japanese as "tsunobikari"). Cake hardness can be roughly measured by a Penetrometer of Japanese Industrial Standard. For powder foundation cosmetics, it is generally known that those having a penetration of 15-25 units give adequate hardness. Ultimately, adequate hardness is determined by actual application of the product using the appropriate applicator for each product type.

Further attempts have been made to formulate pressed powder cosmetic compositions which provide moisturizing effect to the skin.

Patent Laid-open H2-157208 discloses a solid powder cosmetic comprising: (A) 70-99% powder; (B) diglyceride which is in liquid form at room temperature; and (C) polyol moisturizing agent freely soluble with component (B); wherein the weight ratio of component (B) : (C) is within the range of 10:1 - 1:20, and wherein the total of components (B) and (C) is 1-30%.

Patent Laid-open H2-152917 discloses a solid powder cosmetic comprising: (A) 70-99% powder; (B) oil which is in liquid form at room temperature; and (C) liquid surfactant which is insoluble with component

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comprising: (A) 70-99% powder; (B) oil which is in liquid form at room temperature; and (C) liquid surfactant which is insoluble with component (B) at room temperature; wherein the weight ratio of component (B) : (C) is within the range of 20:1 - 1:2, and wherein the total of components (B) and (C) is 1-30%. Moisturizing agent is disclosed as an optional component, however there is no disclosure on the method of incorporating a moisturizing agent.

However, these attempts are not believed to be completely effective for providing a composition comprising an effective amount of moisturizing agent and adequate cake hardness.

Here it has been found that by making a binder base comprising a mixture of a nonionic surfactant and a moisturizing agent, a composition can be made which comprises a high level of moisturizing agent and also retains adequate hardness when pressed into a cake. More specifically, the present composition can comprise up to 15% moisturizing agent without making the obtained pressed cake too hard.

OBJECT OF INVENTION

It is an object of the present invention to provide a pressed powder cosmetic composition which comprises a high level of moisturizing agent and has adequate cake hardness.

It is also an object of the present invention to provide a pressed powder cosmetic composition which has good adhesion and durability on the skin.

It is also an object of the present invention to provide a process for making powder foundations wherein the binder base comprising moisturizing agent is stable and easy to handle.

DETAIL DESCRIPTION

All percentages and ratios are based on weight unless otherwise specified.

SUMMARY

The present invention relates to a pressed cosmetic composition comprising by weight:

- (a) powder colorant; and
- (b) binder base comprising by weight of the cosmetic composition;
 - i) moisturizing agent; and
 - ii) nonionic surfactant which is in liquid or paste form at room temperature;

wherein said binder base meets at least one of the following requirements;

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I) said nonionic surfactant is at least one fifth by weight of said moisturizing agent; or

II) said binder base is a lipophilic gel having a resistivity of at least 10,000 ohm-cm at room temperature.

The present invention also relates to an improved process for making a pressed cosmetic composition comprising the steps of:

- (a) preparing a lipophilic gel binder base by gradually adding under mixing a moisturizing agent into a mixture comprising a nonionic surfactant;
- (b) mixing said lipophilic gel binder base with a powder colorant; and
- (c) pressing the resulting mixture.

POWDER COLORANT

The composition of the present invention comprises 70-99% powder colorant. Powder foundation compositions of the present invention preferably comprise 80-95% powder colorant. Non-limiting examples of powder colorant useful in the present invention are talc, mica, clay, kaolin, zinc oxide, nylon powder, ultramarine, pearl, iron oxide, titanium oxide, titanated mica, calcium carbonate, tar pigments, and other organic pigments. Powder colorant which are surface treated with silicones and its derivatives, metal soaps, fluorine compounds and its derivatives, lecithin and its derivatives, amino acids and its salts, etc. can also be used.

The powder colorant can be incorporated in the composition as is, or as a dispersion in an oily medium, such as any one disclosed below under oily material.

BINDER BASE

The binder base retains the integrity of the pressed powder composition. The composition of the present invention comprises 1-30% of binder base. Powder foundation compositions of the present invention preferably comprise 5-20% binder base. In one embodiment of the present invention, the binder base comprises 2-15% moisturizing agent and 0.4-15% nonionic surfactant, wherein said nonionic surfactant is at least one fifth by weight of moisturizing agent. In the other embodiment of the present invention, the binder base is in the form of a lipophilic gel.

Moisturizing Agent

The moisturizing agent used herein can be one known in the cosmetic art, and provide moisturizing or emollient effect to the skin, and are hydrophilic in nature. The compositions of the present invention comprise 0.1-15%, preferably 2-10% of moisturizing agent. The

moisturizing agent can be a polyhydric alcohol, hyaluronic acid and its salts, an amino acid and its salts, chondroitin sulfuric acid and its salts, lactic acid and its salts, pyroglutamic acid and its salts, uric acid and its salts, and mixtures thereof. A preferred moisturizer is polyhydric alcohol, such as glycerin, diglycerin, triglycerin, ethylene glycol, diethylene glycol, propylene glycol, dipropylene glycol, hexylene glycol, 1,3-butylene glycol, 1,4-butylene glycol, glucose, maltose, sucrose, xylitose, sorbitol, maltitol, malbit, panthenol, hyaluronic acid and its salts, and mixtures thereof. Most preferred is polyhydric alcohol, particularly glycerin, hyaluronic acid, and mixtures thereof. Highly preferred compositions of the present invention comprise at least 3% glycerin.

A moisturizing agent which is solid and practically insoluble in the nonionic surfactant can be introduced as a water solution.

Nonionic Surfactant

The nonionic surfactant is included in the composition to disperse the hydrophilic moisturizing agent in the predominantly powder base of the composition. The composition of the present invention comprises 0.1-15%, preferably 0.4-10% of nonionic surfactant. The level of nonionic surfactant is adjusted according to the level and species of moisturizing agent. For good dispersion of moisturizing agent, the binder base should comprise at least one fifth, preferably at least one half, nonionic surfactant by weight of the moisturizing agent.

The nonionic surfactant in the composition of the present invention should be one which is in liquid or paste form at 25°C. The nonionic surfactant will form a lipophilic gel binder base when moisturizing agent is gradually added thereto under stirring. Combinations of nonionic surfactant of higher and lower melting points can be used, so long as the mixture of the nonionic surfactant is in liquid or paste form at 25°C. The nonionic surfactant should preferably be selected by the skilled artisan in view of the solubility with the moisturizing agent. Nonlimiting examples of nonionic surfactant are polyglycerin fatty acid esters, propylene glycol fatty acid esters, glycerin fatty acid esters, sorbitan fatty acid esters, sugar fatty acid esters, polyoxyethylene sorbitan fatty acid esters, polyoxyethylene sorbit fatty acid esters, polyethylene glycol fatty acid esters, polyoxyethylene castor oils, polyoxyethylene hardened castor oils, polyoxyethylene alkyl ethers, polyoxyethylene phytosterols, polyoxyethylene polyoxypropylene alkyl ethers, polyoxyethylene alkyl phenyl ethers, polyoxyethylene lanolins,

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polyoxyethylene lanolin alcohols, polyoxyethylene beeswax derivatives, polyoxyethylene fatty acid amides, and polyether silicone derivatives.

Polyglycerin fatty acid ester and a combination including polyglycerin fatty acid ester is most preferred. The polyglycerin portion of this type of surfactant is a polymer of 2 or more monomers, usually 2-10 monomers. The fatty acids making the ester can be saturated or unsaturated, straight or branched. Those of natural origin having about 16-18 carbons are most preferred. Among polyglycerin fatty acid esters, preferred is triglyceryl beeswax, triglyceryl cetyl ether, tetraglyceryl cocoate, triglyceryl decyltetradecanol, diglyceryl diisostearate, triglyceryl diisostearate, decaglyceryl diisostearate, diglyceryl dioleate, triglyceryl dioleate, hexaglyceryl dioleate, decaglyceryl dioleate, triglyceryl distearate, hexaglyceryl distearate, decaglyceryl distearate, decaglyceryl trioleate, decaglyceryl heptaoleate, decaglyceryl heptastearate, hexaglyceryl hexaoleate, diglyceryl isostearate, tetraglyceryl isostearate, hexaglyceryl monoisostearate, diglyceryl lanolin alcohol ether, tetraglyceryl lauryl ether, diglyceryl oleate, triglyceryl oleate, tetraglyceryl oleate, hexaglyceryl oleate, diglyceryl oleyl ether, tetraglyceryl oleyl ether, diglyceryl sesquiisostearate, and diglyceryl sesquioleate and mixture thereof. Most preferred is diglyceryl monoisostearate, diglyceryl diisostearate, diglyceryl sesquiisostearate, hexaglyceryl dioleate, and decaglyceryl trioleate, and mixtures thereof.

OPTIONAL COMPONENTS

The compositions of the present invention can contain other optional components.

Water

The composition of the present invention can optionally comprise up to 5% water. Water is not an essential component to the composition of the present invention. However, since certain components such as powder colorant can be hygroscopic, a low level of water can be added to prevent moisture take-up into the composition from either the atmosphere or from the skin. Water can be introduced separately or as a vehicle for other ingredients, such as the moisturizing agents.

Oily Material

The composition of the present invention can optionally comprise up to 20% oily material. An oily material can act as an emollient, and can provide adhesion and durability properties to the cosmetic. The oily material is preferably in liquid or paste form at room temperature. Said

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oily material can be hydrocarbon oil, natural oil, fatty alcohol, fatty acid ester, silicone oil, and paste wax and mixtures thereof.

Non-limiting examples of the hydrocarbons are liquid paraffin, squalane, liquid petrolatum, mineral oil, and liquid polybutene.

The natural oil used herein typically is a mixture of saturated and unsaturated fatty acid. Non-limiting examples of natural oil derived from plants include almond oil, olive oil, sesame oil, safflower oil, avocado oil, cottonseed oil, jojoba oil, castor bean oil, castor oil, rapeseed oil, soybean oil, palm kernel oil, coconut oil, hydrogenated vegetable oil, and cocoa butter. Non-limiting examples of natural oil derived from animal sources include mink oil and egg yolk oil.

Non-limiting examples of fatty alcohol useful in the present invention are isostearyl alcohol, lanolin alcohol, oleyl alcohol, hexadecyl alcohol, octyldodecanol alcohol, linoleyl alcohol, linolenyl alcohol, and arachidyl alcohol.

Fatty acid can be natural or synthetic, saturated, unsaturated, linear, or branched. Non-limiting examples of fatty acid useful in the present invention are adipic, caprylic, capric, isostearic, linoleic, ricinoleic, oleic, elaidic and erucic acid.

Non-limiting examples of the fatty acid ester are cetyl ricinoleate, cetyl oleate, cetyl octanoate, cetyl acetate, glyceryl trioctanoate, isopropyl lanolate, isopropyl linoleate, isopropyl myristate, isopropyl palmitate, isopropyl oleate, isopropyl stearate, ethyl lactate, ethyl glutamate, ethyl laurate, ethyl linoleate, ethyl methacrylate, ethyl myristate, ethyl palmitate, diisopropyl adipate, octyl dodecyl myristate, octyl palmitate, octyl isopelargonate, octyl dodecyl lactate, tridecyl isononanoate, isotridecyl isononanoate, hexadecyl stearate, oleyl oleate, isononyl isononanoate, isostearyl myristate, dipenta-erythritol ester, neopentyl glycol dioctanoate, and di(capryl/capric acid) propylene glycol and mixtures thereof. Other suitable esters include triglycerides such as caprylic triglycerides, capric triglyceride, isostearic triglyceride, adipic triglyceride and cholesterol derivatives such as cholesteryl oleate.

Non-volatile, straight, and branched silicone oil such as dimethicone and phenyl dimethicone is also useful.

Other Optional Components

Other optional components include antioxidants, preservatives, liquid and paste pigments, antiinflammatory agents, astringents, pH buffers, perfumes, ultraviolet and infrared screening agents, amphoteric

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and anionic surfactants such as lecithins and phosphates, solid amorphous lipids, vitamins, nutrients, and skin conditioning agents.

Antioxidants and preservatives can be, and usually will be formulated in the composition of the present invention. Useful antioxidants and preservatives are tocopherol, dibutylhydroxy toluene, methylparaben, and propylparaben. These components are typically individually present at levels less than 1 %.

Useful skin conditioning agents are beta-glycyrrhetic acid and its derivatives, vegetation extracts, allantoin, collagen, and extract and treated elastin fibers.

PROCESS

The present composition is made by first preparing a binder base by mixing nonionic surfactant and moisturizing agent, and optionally an oily material.

In one embodiment of the present invention, the binder base comprises 2-15% moisturizing agent and 0.4-15% nonionic surfactant, wherein said nonionic surfactant is at least one fifth by weight of moisturizing agent. With this amount of nonionic surfactant against moisturizing agent, the obtained binder base can hold a substantial ratio of moisturizing agent, up to 15% to the composition. In this embodiment, moisturizing agent, nonionic surfactant, and other components of the binder base can be mixed in any convenient manner to form a binder base which is suitable for spraying onto or kneading with the powder colorants.

In another embodiment of the present invention, the binder base comprises 0.1-15% moisturizing agent and nonionic surfactant respectively, and is in the form of a lipophilic gel. Such lipophilic gel binder base can be made by gradually adding the moisturizing agent into the nonionic surfactant or mixture of nonionic surfactant and optional oily material. Mixing can be done by a preferred mixer in the cosmetic art such as Anchor-type, Daysolver, Homomixer, and combinations thereof. The lipophilic gel binder base herein obtained has a lipophilic gel structure having a resistivity of at least about 10,000 $\Omega \cdot \text{cm}$ at room temperature. Resistivity can be measured, for instance by a resistivity measurer Type MR-45, Class 2.5 (Okasawa-denki). Such lipophilic gel structure is different from water-in-oil or oil-in-water emulsions. This lipophilic gel binder base is stable in phase, and does not separate for at least a few hours at room temperature. This is important and useful, because this lipophilic gel binder base does not

have to be repeatedly mixed while spraying onto or kneading with the powder colorants. Further, since this lipophilic gel binder base is homogeneous, a pressed powder product of uniform composition is obtained, thus providing an adequate hardness to the pressed cake. It has been found that when components of the binder base are not uniform and/or are separated in phase, the obtained cake tends to become too hard. The components of the lipophilic gel binder base are preferably made in one of the following ways:

- i) gradually adding moisturizing agent into nonionic surfactant;
- ii) gradually adding moisturizing agent into nonionic surfactant, followed by adding obtained mixture into oily material; or
- iii) mixing nonionic surfactant and oily material, followed by gradually adding moisturizing agent into obtained mixture.

The binder base thus obtained by either embodiment of the present invention is then mixed with the powder colorant. This can be done by spraying the binder base onto the powder colorant while tumbling. The powder colorants are preferably pulverized before spraying on the binder base. (In the context of the present invention, "pulverizing" particularly pertains to deagglomerating particles to their original size). If the binder base is too viscous to handle by spraying, the binder base can be kneaded with the powder colorants, and pulverized afterwards. After the powder colorant and binder base are mixed, the obtained composition is de-agglomerated and sifted to obtain consistent particle size, poured into a mold, and pressed into a cake. The whole composition making process, namely; preparing the binder base, spraying, pulverizing and pressing; can be done at room temperature.

Any optional components are incorporated in the composition of the present invention by dissolving or dispersing in a suitable phase in which it has affinity. Oily material can be used to make the lipophilic gel binder base, or otherwise, mixed with the powder colorants independently. Oily material can be independently mixed before or after the binder base (comprising moisturizing agent and nonionic surfactant) is mixed with powder colorant. Other lipophilic optional components can be dissolved or dispersed with powder colorants or liquid oils. Water and other hydrophilic optional components can be dissolved or dispersed with moisturizing agents.

EXAMPLES

The following examples illustrate the compositions of the present invention, but are not intended to be limiting thereof. All percentages and ratios are based on weight unless otherwise specified.

EXAMPLE 1

[Powder Foundation]

TABLE 1

COMPONENTS		AMOUNT (%)
A	MICA	56.2
	METHICONE COATED MICA	15.0
	TITANIUM DIOXIDE	10.0
	NYLON-12	5.0
	YELLOW IRON OXIDE	1.5
	BLACK IRON OXIDE	0.25
	RED IRON OXIDE	0.4
	METHYLPARABEN	0.1
B	PROPYLPARABEN	0.05
	POLYGLYCERYL-2 DIISOSTEARATE	5.5
C	GLYCERIN	5.0
	WATER	1.0

The components shown in Table 1 are prepared as follows:

Group C is gradually added to Group B under stirring with an Anchor mixer to make a lipophilic gel binder base. Separately, Group A is mixed in a Ribbon mixer until homogeneous, followed by pulverizing. The obtained lipophilic gel binder base is sprayed onto the pulverized Group A while tumbling. The finally obtained mixture is deagglomerated, sifted, and pressed into a solid product.

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EXAMPLE 2

[Eyeshadow]

TABLE 2

	COMPONENTS	AMOUNT (%)
A	TALC	42.85
	ULTRAMARINE	1.50
	RED IRON OXIDE	1.00
	D&C RED #30	0.50
	METHYLPARABEN	0.10
B	KAOLIN	15.00
	MAGNESIUM CARBONATE	1.00
	ZINC STEARATE	10.00
	TITANIUM DIOXIDE	5.00
	TITANATED MICA	10.00
C	MINERAL OIL	4.00
	LANOLIN	1.00
	PROPYLPARABEN	0.05
D	SORBITAN SESQUIOLEATE	1.00
	DECAGLYCERIN TRIOLEATE	2.00
E	GLYCERIN	3.00
	BUTYLENE GLYCOL	2.00

The components shown in Table 2 are prepared as follows:

Group E is gradually added to Group D under mixing with an Anchor mixer to make a lipophilic gel binder base. Separately, Group A is combined, followed by further mixing with Group B in a Ribbon mixer, and pulverized together to make a powder colorant mixture. Group C is mixed and sprayed onto the powder colorant mixture while tumbling. The lipophilic gel binder base is added to the obtained mixture and kneaded homogeneously. The finally obtained mixture is then treated as in Example 1.

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EXAMPLE 3

[Blusher]

Table 3

COMPONENTS		AMOUNT (%)
A	TALC	56.35
	KAOLIN	20.00
	TITANIUM DIOXIDE	4.00
	ZINC STEARATE	5.00
	CORN STARCH	5.00
	YELLOW IRON OXIDE	0.50
	RED IRON OXIDE	2.30
	BLACK IRON OXIDE	0.60
	METHYLPARABEN	0.10
B	MINERAL OIL	3.00
	PROPYLPARABEN	0.05
	DIGLYCERIN OLEATE	1.00
	PERFUME	0.10
C	GLYCERIN	2.00

The components shown in Table 3 are prepared as follows:

Group C is gradually added to Group B under mixing with an Anchor mixer to make a lipophilic gel binder base. Separately, Group A is mixed in a Ribbon mixer until homogeneous, followed by pulverizing. The obtained lipophilic gel binder base is sprayed onto the treated Group A while tumbling. The finally obtained mixture is then treated as in Example 1.

EXAMPLE 4

[Face Powder]

TABLE 4

COMPONENTS		AMOUNT (%)
A	TALC	42.85
	YELLOW IRON OXIDE	0.20
	RED IRON OXIDE	0.10
	METHYLPARABEN	0.10
B	KAOLIN	20.00
	MAGNESIUM STEARATE	3.00
	ZINC OXIDE	15.00
	TITANIUM DIOXIDE	5.00
	CORN STARCH	2.00
C	MINERAL OIL	3.00
	LANOLIN	1.50
	PROPYLPARABEN	0.05
D	TETRAGLYCERIN ISOSTEARATE	0.50
E	BUTYLENE GLYCOL	2.00
F	WATER	1.00
	SODIUM HYALURONATE	0.10

The components shown in Table 4 are prepared as follows:

Group F is homogeneously dissolved. Group E is added to Group F and homogeneously dissolved. The obtained mixture, followed by previously mixed Group C are gradually added to Group D under mixing to make a lipophilic gel binder base. Separately, Group A and Group B are mixed separately, mixed together, and pulverized to make a powder colorant mixture. The above obtained lipophilic gel binder base is sprayed onto the treated powder colorant mixture while tumbling. The finally obtained mixture is then treated as in Example 1.

EXAMPLE 5

(Lip Powder)

TABLE 5

COMPONENTS		AMOUNT (%)
A	TALC	16.75
	TITANATED MICA	50.00
	SILICA	5.00
	D&C RED #30	2.00
	METHYLPARABEN	0.10
B	PHENYL DIMETHICONE	10.00
	LANOLIN	8.00
	TOCOPHEROL	0.10
	PROPYLPARABEN	0.05
C	DIGLYCERIN TRIISOSTEARATE	3.00
D	GLYCERIN	2.00
	SORBITOL	3.00

The components shown in Table 5 are prepared as follows:

Group D is gradually added to Group C under mixing with a Daysolver to make a lipophilic gel binder base. Separately, Group A is mixed in a Ribbon mixer and pulverized. The above obtained lipophilic gel binder base is sprayed onto treated Group A while tumbling. The obtained mixture is further mixed with previously mixed Group C. The finally obtained mixture is then treated as in Example 1.

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What is Claimed is:

1. A pressed cosmetic composition comprising by weight:
 - (a) 70-97.6% powder colorant; and
 - (b) 2.4-30% binder base comprising by weight of the cosmetic composition:
 - i) 2-15% moisturizing agent; and
 - ii) 0.4-15% nonionic surfactant which is in liquid or paste form at 25°C;wherein said nonionic surfactant is at least one fifth by weight of said moisturizing agent.
2. A pressed cosmetic composition comprising:
 - (a) 70-99% powder colorant; and
 - (b) 1-30% lipophilic gel binder base having a resistivity of at least 10,000 $\Omega \cdot \text{cm}$ at room temperature comprising by weight of the cosmetic composition:
 - i) 0.1-15% moisturizing agent; and
 - ii) 0.1-15% nonionic surfactant which is in liquid or paste form at 25°C.
3. A pressed cosmetic composition according to Claim 1 or 2 wherein said moisturizing agent is selected from the group consisting of polyhydric alcohol, hyaluronic acid, and mixtures thereof.
4. A pressed cosmetic composition according to Claim 3 wherein said nonionic surfactant is polyglycerin fatty acid ester.
5. A pressed cosmetic composition according to Claim 4 comprising at least 3% glycerin.
6. A pressed cosmetic composition according to Claim 1 or 2 wherein said nonionic surfactant is at least one half by weight of said moisturizing agent.

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7. A pressed powder foundation composition according to Claim 3 comprising 80-95% powder colorant.
8. A process for making a pressed cosmetic composition comprising the steps of:
 - (a) preparing a lipophilic gel binder base by gradually adding under mixing a moisturizing agent into a mixture comprising a nonionic surfactant;
 - (b) mixing said lipophilic gel binder base with a powder colorant; and
 - (c) pressing the resulting mixture.

INTERNATIONAL SEARCH REPORT

 International Application No.
 PCT/US 94/05310

 A. CLASSIFICATION OF SUBJECT MATTER
 IPC 5 A61K7/02

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

 Minimum documentation searched (classification system followed by classification symbols)
 IPC 5 A61K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US,A,4 980 157 (MERCADO ET AL.) 25 December 1990	
A	JP,A,2 157 208 (KAO) 18 June 1990 cited in the application	
A	JP,A,2 152 917 (KAO) 12 June 1990 cited in the application	

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☒ Patent family members are listed in annex.

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 Tel. (+31-70) 340-2040, Tx. 31 631 epo nl,
 Fax (+31-70) 340-3016

Authorized officer

Klaver, T

INTERNATIONAL SEARCH REPORT

information on patent family members

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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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